

Nodeless superconductivity in quasi-one-dimensional Nb₂PdS₅: a μ SR study

P. K. Biswas,^{1,*} H. Luetkens,¹ Xiaofeng Xu,² J. H. Yang,² C. Baines,¹ A. Amato,¹ and E. Morenzoni¹

¹*Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland*

²*Department of Physics and Hangzhou Key Laboratory of Quantum Matters,
Hangzhou Normal University, Hangzhou 310036, China*

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Muon spin relaxation and rotation (μ SR) measurements have been performed to study the superconducting and magnetic properties of Nb₂PdS₅. Zero-field μ SR data show that no sizeable spontaneous magnetization arises with the onset of superconductivity in Nb₂PdS₅ which indicates that the time reversal symmetry is probably preserved in the superconducting state of this system. A strong diamagnetic shift is observed in the transverse-field (TF) μ SR data practically ruling out a dominant triplet-pairing superconducting state in Nb₂PdS₅. The temperature dependence of magnetic penetration depth evidences the existence of a single s -wave energy gap, $\Delta(0)$ with a gap value of 1.07(4) meV at zero temperature. The ratio $\Delta(0)/k_B T_c = 2.02(9)$ indicates that Nb₂PdS₅ should be considered as a moderately strong-coupling superconductor. The magnetic penetration depth at zero temperature is 785(20) nm, indicating a very low superfluid density consistent with the quasi-one-dimensional nature of this system.

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The recent discovery of superconductivity in the quasi-one-dimensional (Q1D) material Nb₂PdS₅ with a T_c of 6.6 K has attracted interest due to its remarkably large and anisotropic upper critical field ($H_{c2} > 370$ kOe along the b -axis) [1, 2], far above its Pauli paramagnetic limit ($H_p = 121$ kOe). Layered superconductors generally exhibit large H_{c2} values, which often significantly exceed the paramagnetic limit. Examples include high- T_c cuprates [3] and Fe-based superconductors [4–7]. There are several circumstances under which H_{c2} may exceed the Pauli limit. These are strong spin-orbit scattering [13, 14], multiband effect [8, 9], spatially inhomogeneous Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) state [10–12] and spin-triplet pairing [13, 14].

Nb₂PdS₅ crystallizes in the low-dimensional monoclinic space group $C2/m$ [1]. Density functional theory calculations show that the Fermi surface (FS) is composed of Quasi-2D sheets of hole character and strongly warped Quasi-1D sheets with both electron and hole characters [1]. The large H_{c2} in this system was tentatively ascribed to multiband effects or spin-triplet pairing [1, 2]. Recently it has been shown that H_{c2} of this system can be modified in a tunable fashion by chemical doping on the Pd chains with elements of varying mass numbers and hence by altering the spin-orbit coupling on the Pd sites [15]. It was also suggested that superconductivity in Nb₂PdS₅ may appear in proximity to a magnetic instability. Very recent specific heat measurements on this material however show that the size of the heat capacity jump at T_c is smaller than the BCS value and that a nodeless gap opens at the Fermi surface [16]. All these experimental data are far from conclusive and the exact nature of its unconventional SC phases has yet to be resolved. To gain further information about the superconducting properties

and magnetic state of this transition metal-chalcogenide-based Q1D compound, we have performed a detailed μ SR study of this system. Our zero-field (ZF)- μ SR results find no evidence of any sizeable magnetism in the superconducting state of Nb₂PdS₅. Transverse-field (TF)- μ SR results show that, at low temperature, the superfluid density $\rho_s \propto \lambda^{-2}$ (λ is the magnetic penetration depth) becomes temperature independent, which is consistent with a fully gapped superconducting state. $\rho_s(T)$ can be well fitted with a single-gap s -wave model with a gap value of 1.07(4) meV at absolute zero temperature. This results in a gap to T_c ratio of 2.02(9) suggesting a strong coupling of the superconducting charge carriers. The absolute value of the magnetic penetration depth is determined to be $\lambda(0) = 785(20)$ nm.

Polycrystalline samples of Nb₂PdS₅ were grown by a solid-state reaction method [1] and characterized as described in Ref. [16]. The resistivity (with a standard four-probe technique) and DC magnetization measurements were performed in a Quantum Design Physical Property Measurement System (PPMS) and Quantum Design Magnetic Property Measurement System (MPMS), respectively. The TF- and ZF- μ SR experiments were carried out in the LTF instrument at the π M3 beam line of the Paul Scherrer Institute (Villigen, Switzerland). The sample was cooled to the base temperature in zero field for the ZF- μ SR experiments and in 695.7 Oe for the TF- μ SR experiments. Typically ~ 12 million muon decay events were collected for each spectrum. The ZF- and TF- μ SR data were analyzed by using the free software package MUSRFIT [17].

Figure 1 shows the temperature dependence of the resistivity of Nb₂PdS₅ under zero magnetic field. Zero-resistivity can be observed at 6.05 K. The inset shows the temperature variation of the DC magnetic susceptibility

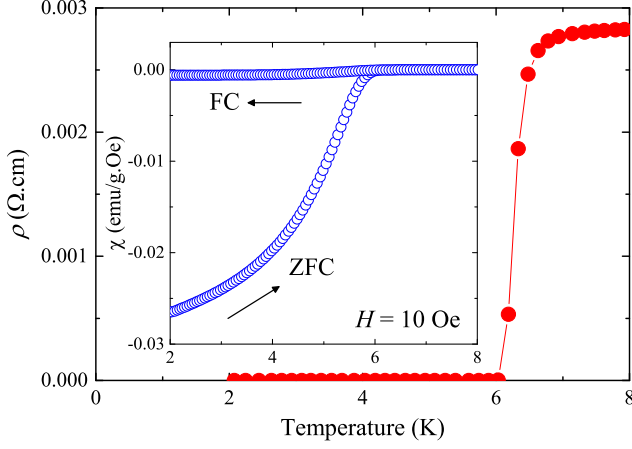


FIG. 1: (Color online) Resistivity vs. temperature curve for Nb_2PdS_5 . The inset shows the temperature variation of the DC magnetic susceptibility curves of Nb_2PdS_5 in ZFC and FC modes in an applied field of 10 Oe.

of Nb_2PdS_5 in zero-field-cooled (ZFC) and field-cooled (FC) modes in an applied field of 10 Oe. ZFC susceptibility shows a diamagnetic transition (onset) at around 6 K, a value very close to the zero-resistivity T_c .

Figure 2(a) compares the ZF- μSR signals collected above and below T_c which are practically identical. The ZF- μSR data can be well described using a combination of Gaussian and Lorentzian Kubo-Toyabe (KT) relaxation functions [18],

$$A(t) = A(0) \left\{ \frac{1}{3} + \frac{2}{3} (1 - a^2 t^2 - \Lambda t) \exp \left(-\frac{a^2 t^2}{2} - \Lambda t \right) \right\} + A_{\text{bg}}, \quad (1)$$

where $A(0)$ is the initial asymmetry of the sample signal, A_{bg} is the background signal, a and Λ are the muon spin relaxation rates due to randomly oriented Nb and diluted ^{105}Pd nuclear moments, respectively. The nuclear moment of S is negligible compare to the others. The KT relaxation function is of Gaussian type when the nuclear moments are densely populated in a lattice. A Lorentzian type of relaxation is expected for weak and diluted nuclear or electronic moments. For this material, Nb has a reasonably strong nuclear magnetic moment which is densely populated and therefore produce Gaussian type field distribution. On the other hand, Pd has several isotopes with only one of them carrying a nuclear moment. This isotope has a natural abundance of only 22% and therefore most probably causes a field distribution similar to a Lorentzian. The fits to the ZF- μSR signals using Eq. 1 yield, $a = 0.16(2) \mu\text{s}^{-1}$, $\Lambda(19\text{mK}) = 0.079(4) \mu\text{s}^{-1}$, and $\Lambda(8\text{K}) = 0.065(3) \mu\text{s}^{-1}$. Comparing the two ZF- μSR signals, collected above and below T_c , we see a marginally small increase of damping rate corresponds to a magnetic field width increase of 0.16 G at low temperature. One

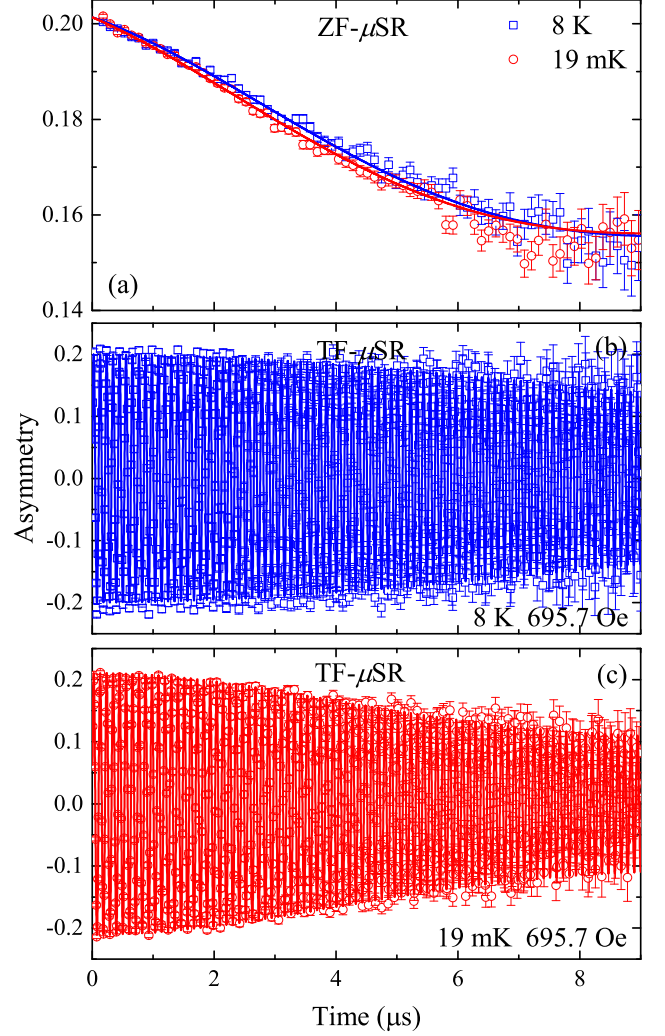


FIG. 2: (Color online) (a) ZF- μSR spectra of Nb_2PdS_5 taken at 8 K (above T_c) and 19 mK (below T_c). The solid lines are fits to the data using Eq. 1. (b) and (c) show the TF- μSR time spectra, collected both above and below T_c in an applied field of 695.7 Oe. The solid lines are fits to the data using Eq. 2.

could argue that this additional contribution might be related to the spontaneous magnetic moments associated with a time-reversal-symmetry (TRS) breaking pairing state in Nb_2PdS_5 [19–27]. However, it is not sizeable enough to make such a claim for Nb_2PdS_5 . Further studies are in progress to explore the presence of TRS breaking spontaneous magnetic moments in this material. In the absence of TRS breaking fields, an alternative explanation for such a small field is the presence of a small amount of magnetic impurities in the sample.

Figure 2(b) and (c) show the TF- μSR time spectra collected at 8 K and 19 mK in a transverse field of 695.7 Oe. The data obtained in the normal state show a very weak local relaxation of the μSR time spectra. In this case the local field probed by the muons is essentially the applied

field. The signal is only slightly damped by the nuclear moments contribution. By contrast, the data collected in the superconducting state shows a more pronounced damping due to the inhomogeneous field distribution generated by the formation of a vortex lattice in Nb₂PdS₅. The TF- μ SR time spectra were analyzed using a Gaussian damped precession signal:

$$A^{TF}(t) = A(0) \exp(-\sigma^2 t^2 / 2) \cos(\gamma_\mu \langle B \rangle t + \phi) \exp(-\alpha t) + A_{bg}(0) \cos(\gamma_\mu B_{bg} t + \phi), \quad (2)$$

where $A(0)$ and $A_{bg}(0)$ are the initial asymmetries of the sample and background signals, $\gamma_\mu/2\pi = 13.55$ kHz/G is the muon gyromagnetic ratio [28], $\langle B \rangle$ and B_{bg} are the internal and background magnetic fields, ϕ is the initial phase of the muon precession signal, σ is the Gaussian muon spin relaxation rate, and α is the exponential relaxation rate due to weak and diluted nuclear or electronic moments. The background signal mainly originates from muons hitting the silver sample holder, where the relaxation rate is negligibly small. A global fit to the TF- μ SR signals using Eq. 2 yield $\alpha = 0.031(3) \mu s^{-1}$.

Figure 3 (a) shows the temperature dependence of σ of Nb₂PdS₅ for an applied field of 695.7 Oe. The temperature dependence of the internal magnetic field at the muon site with the expected diamagnetic shift below T_c is shown in Fig. 3 (b). The dashed lines are guides to the eye only.

The superconducting contribution to σ is obtained by subtracting the nuclear moment contribution (measured above T_c) as $\sigma_{sc}^2 = \sigma^2 - \sigma_{nm}^2$. In an isotropic type-II superconductor with a hexagonal Abrikosov vortex lattice described by Ginzburg-Landau theory, the magnetic penetration depth λ is related to σ_{sc} by the Brandt equation [29]:

$$\sigma_{sc} [\mu s^{-1}] = 4.854 \times 10^4 \left(1 - \frac{H}{H_{c2}}\right) \left[1 + 1.21 \left(1 - \sqrt{\frac{H}{H_{c2}}}\right)^3\right] \frac{1}{\lambda_{nm}^2}, \quad (3)$$

where H and H_{c2} are the applied and upper critical field, respectively. The temperature dependence of $\lambda^{-2}(T)$ was calculated by using Eq. 3 and $H_{c2}(T)$ from Ref. [16]. Fig. 4 shows the temperature dependence of λ^{-2} which is proportional to the effective superfluid density, $\rho_s \propto \lambda^{-2}$. As Fig. 4 illustrates, ρ_s is very nearly constant below $T_c/3 \approx 2$ K. This possibly suggests the absence of low-lying excitations and is indicative of a nodeless superconducting gap at the Fermi surface below T_c in Nb₂PdS₅. This is further verified by a good fit to the $\lambda^{-2}(T)$ data with a single-gap BCS s -wave model (solid line in Fig. 4) [30, 31]:

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = 1 + 2 \int_{\Delta(T)}^{\infty} \left(\frac{\partial f}{\partial E}\right) \frac{E dE}{\sqrt{E^2 - \Delta(T)^2}}. \quad (4)$$

Here $\lambda^{-2}(0)$ is the zero-temperature value of the magnetic penetration depth and $f = [1 + \exp(E/k_B T)]^{-1}$ is

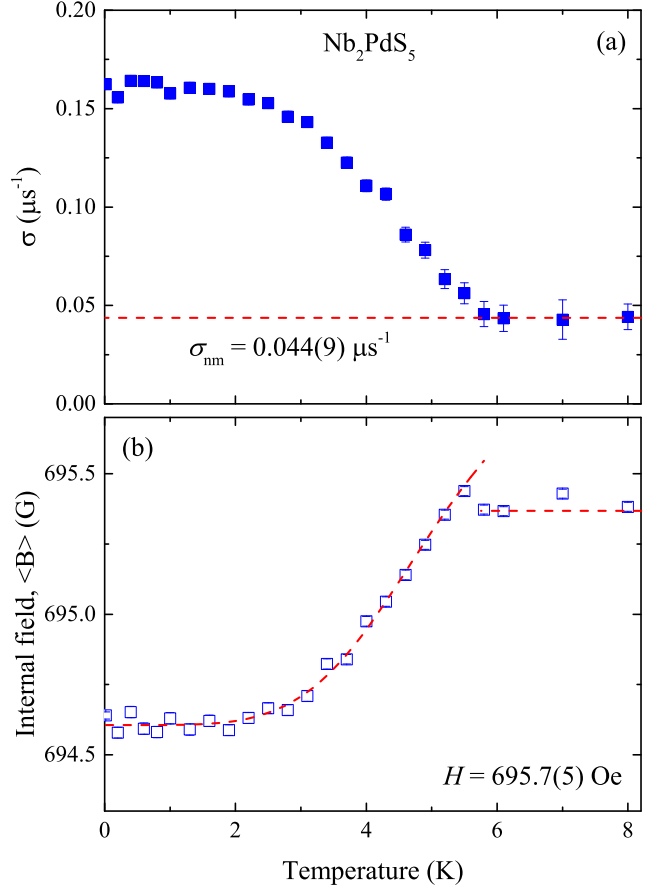


FIG. 3: (Color online) (a) Temperature dependence of the muon depolarization rate σ of Nb₂PdS₅ collected in an applied magnetic field of 695.7 Oe. (b) Typical diamagnetic shift of the internal field experienced by the muons below T_c . The dashed lines are guides to the eye only.

the Fermi function. The BCS temperature dependence of the superconducting gap function is approximated as [32]

$$\Delta(T) = \Delta(0) \tanh \left\{ 1.82 \left[1.018 \left(\frac{T_c}{T} - 1 \right) \right]^{0.51} \right\} \quad (5)$$

where $\Delta(0)$ is the gap magnitude at zero temperature. The fit yields $T_c = 6.2(2)$ K, $\lambda(0) = 785(20)$ nm, and $\Delta(0) = 1.07(4)$ meV. The gap to T_c ratio $\Delta(0)/k_B T_c = 2.02(9)$ is higher than the BCS value of 1.76, suggesting that Nb₂PdS₅ is a moderately strong-coupling superconductor. It is noteworthy to mention that, though the BCS model is appropriate for a weak-coupling superconductor, standard BCS theory has been widely used in the past to determine electronic parameters even for systems which do not fulfill all the strict criteria of a BCS superconductor. In exotic superconductors, such as cuprates and Fe-based systems, the value and symmetry of the gap obtained from μ SR using the BCS expression are in many cases in good agreement with values obtained by other spectroscopic methods such as ARPES [33]. We

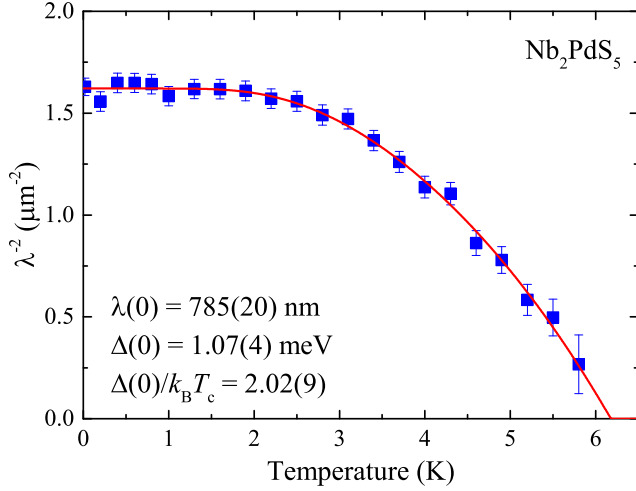


FIG. 4: (Color online) The temperature dependence of $\lambda^{-2}(T)$. The solid line is a fit to the data with the weak-coupling BCS model.

add the value of $\lambda(0)$ for Nb_2PdS_5 as a red star to the Uemura plot, shown in Fig. 5, which shows the scaling relation between $\lambda^{-2}(0)$ and T_c for unconventional superconductors [34]. The value of $\lambda^{-2}(0)$ places Nb_2PdS_5 well inside the broad line for cuprates and iron-based superconductors on the Uemura plot.

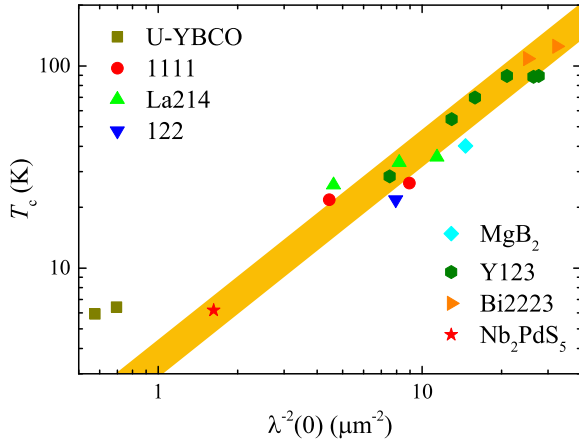


FIG. 5: (Color online) The Uemura plot for high T_c cuprates, MgB_2 and Fe-based superconductors with the data points taken from Ref. [35–37]. The red star represents the data for Nb_2PdS_5 obtained in this work.

Using $H_{c2}(0) = 176(2)$ kOe (estimated by fitting the $H_{c2}(T)$ data of Ref. [16] with a Ginzburg-Landau (GL) model [30]) and its relation with the coherence length ξ , $\left(\mu_0 H_{c2} = \frac{\phi_0}{2\pi\xi^2}\right)$, we calculate $\xi = 4.3(1)$ nm at 0 K. Using the values of λ and ξ , we calculate the GL pa-

rameter, $\kappa = \frac{\lambda}{\xi} \cong 183$. The large value of κ indicates that Nb_2PdS_5 is an extreme type-II superconductor. For completeness, by combining the value of ξ and our measured value of λ , we calculate the value of lower critical field, H_{c1} using the expression [29]:

$$\mu_0 H_{c1} = \frac{\phi_0}{4\pi\lambda^2} \left(\ln \frac{\lambda}{\xi} + 0.5 \right) \quad (6)$$

and obtain $H_{c1}(0) = 14(1)$ Oe.

In conclusion, μSR measurements have been performed on superconducting Nb_2PdS_5 . ZF- μSR results do not find evidence of any sizeable spontaneous magnetization in Nb_2PdS_5 . TF- μSR results show that the superfluid density becomes practically temperature independent below $T_c/3 \approx 2$ K, which is consistent with a fully gapped superconducting state. $\rho_s(T)$ can be well described within the single-gap BCS s -wave scenario with $\Delta(0) = 1.07(4)$ meV. The magnetic penetration depth was estimated to $\lambda(0) = 785(20)$ nm, which is significantly large but consistent with the $\lambda(0)$ values of other low-dimensional superconductors [38–43]. For layered superconductors, such as cuprates [44] and iron chalcogenides [45], it has been observed in the past that the penetration depth increases with increasing layer separation, i.e. when the superfluid density goes from 3D to 2D nature. The same is true when superfluid density becomes Q1D in nature and explains the finding of such a large $\lambda(0)$ in this system. There is no signature of multiple gaps, as it may be expected from the multiband structure of this material possibly indicating that the gaps have a similar magnitude. The value of the gap to T_c ratio, 2.02(9) is higher than the BCS value of 1.76 and suggests that Nb_2PdS_5 is a moderately strong-coupling superconductor. This is probably partially responsible for such a large H_{c2} along the b -axis, similar to heavy-fermion superconductors, such as UBe_{13} [46]. The value of $\lambda^{-2}(0)$ places Nb_2PdS_5 well inside the broad line for unconventional superconductors in a Uemura plot. The results presented here will provide a reference point for further μSR studies on single crystals at high magnetic field, applied both parallel and perpendicular to the principle b -axis.

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* Corresponding author: pabitra.biswas@psi.ch

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